

Initial testing of a low pressure permeator for tritium processing

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1. Introduction

Early tritium processing system operations led to relatively large environmental releases of tritium. This led to the inclusion of large oxidation/adsorption systems between tritium systems and the facility stack so that tritium would be collected as water. While this has been an improvement, it has resulted in large amounts of tritiated water waste. Also it has been recognized for some time that there are cases where inert gases such as Ar, He and N₂ are mixed with small amounts of tritium. If this gas is processed with oxidation/adsorption, the tritium, which is in the useful T₂ form, is converted to the useless T₂O form. Thus, there is a need for a system, which removes tritium from inert gases with high conversion so that the inert gas is ready for release to the environment, and that recovers tritium as T₂.

A system, which could meet this requirement, is one based on getters. The hydrogen isotopes are collected as metal hydride and the inert

passes through. However, such systems are susceptible to rapid consumption of the metal if air or O₂ is present.

2. A promising new system

Thus, there is interest in a new system for tritiated inert gas processing. It has long been known that Pd/Ag permeators can effectively separate hydrogen isotopes (Q₂) from other gases. Practically, Pd/Ag is only permeable to Q₂. A gas mixture is fed to one side of a Pd/Ag membrane and a vacuum is applied to the other side of the membrane. The difference in hydrogen isotope partial pressure across the membrane is the driving force, which generates a flow of Q₂ from one side to the other. To date such devices have been used extensively to practically reduce the Q₂ partial pressure in a gas down to around 1 Torr. This is insufficient for stacking the resulting gas. However, previous work [1] suggested that a Pd/Ag permeator could be used effectively at much lower partial pressures. Thus, the present work was undertaken to explore this idea in a system which could be scaled to a practical device.

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3. Experimental configuration

3.1. Permeator

A very simple, single tube permeator was selected to test the principle of a low pressure permeator. The Pd/Ag tube was 27.7 cm (10.9 in.) long and 0.328 cm (0.125 in.) outer diameter with a 0.0086 cm (0.003 in.) thick wall. Both ends of this tube were brazed to 0.635 cm (0.25 in.) stainless steel tubes. As shown in Fig. 1, this assembly was housed within a 66.0 cm (26 in.) long \times 10.2 cm (4 in.) diameter stainless steel shell. A turbomolecular pump backed by a diaphragm pump was used to maintain vacuum within the shell. One of the stainless steel tubes was used to admit mixtures of inert and H₂ (as a model for T₂) to the inside of the Pd/Ag membrane while the other tube was used to exhaust the unpermeated gas

(retentate) outside the shell. The entire permeator assembly was contained within a tube furnace, which maintained the assembly temperature at the desired value.

3.2. Test stand

The permeator experiments were performed with the setup shown in Fig. 2. Mass flow controllers were used to prepare mixtures of H₂ and either Ar or N₂ supplied from gas cylinders. The pressure of the feed and retentate were measured with Baratron transducers. The permeate vacuum was measured with a cold cathode gauge. The retentate hydrogen concentration was measured with an MTI micro gas chromatograph using a 10 m molecular sieve column and a thermal conductivity detector (TCD). Calibration showed that the TCD could detect down to 0.3 ppm H₂.

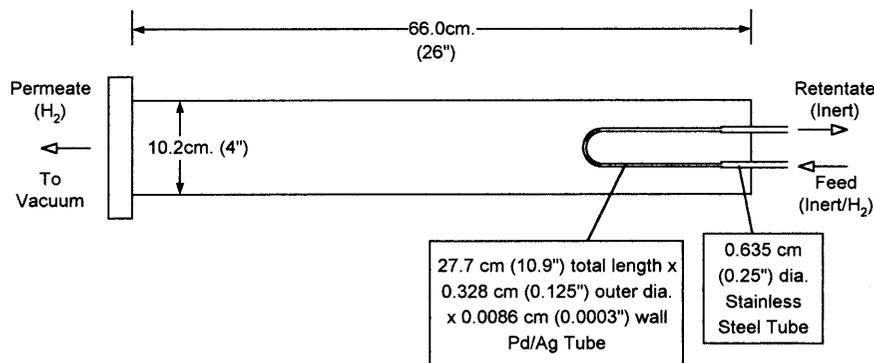


Fig. 1. Mechanical configuration of the permeator.

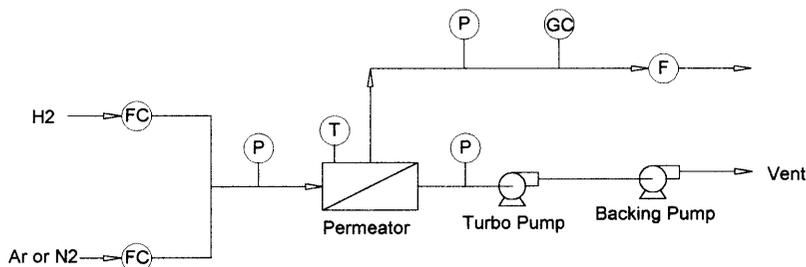


Fig. 2. Experimental set-up. (FC, flow controller, P, pressure, T, temperature, GC, gas chromatograph, F, flow meter).

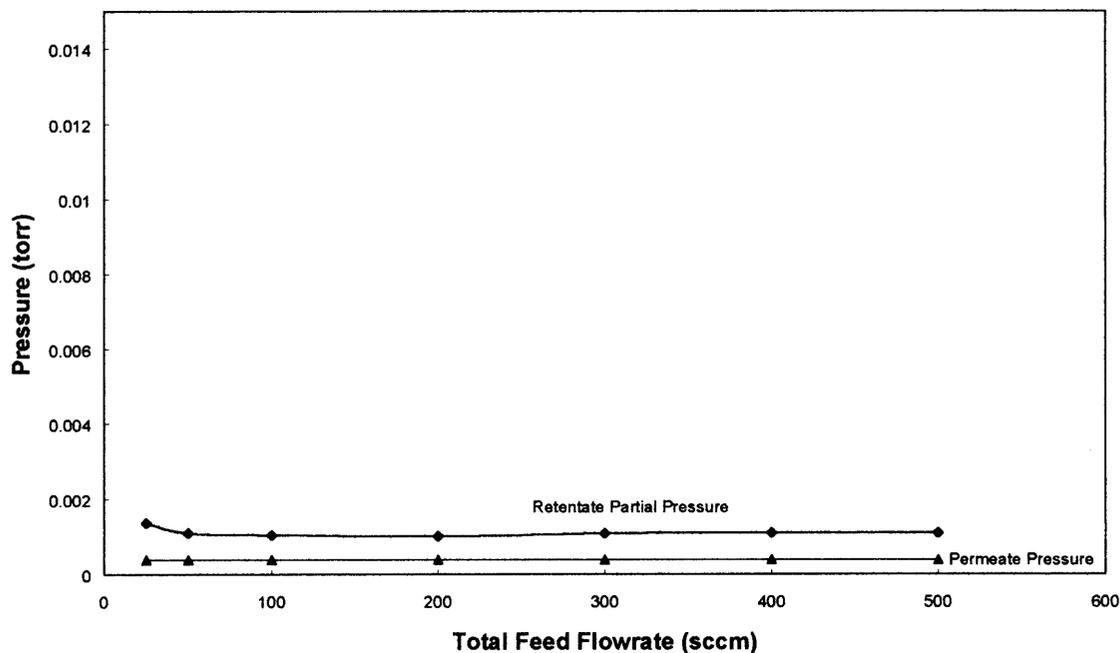


Fig. 3. Results at 450°C and 5 sccm H₂.

3.3. Procedure

The permeator was heated to 350, 400 or 450°C and the pumping system was turned on. A fixed flowrate of H₂ (1, 5, 25, 50 or 75 sccm) was maintained during an experimental run while the flowrate of inert was varied between about 10 and 500 sccm. For each flowrate combination the pressures, temperature, flowrates and gas composition were recorded when conditions reached steady state. The procedure using a fixed H₂ flowrate was chosen so that a fixed load was applied to the turbomolecular pump, resulting in a fixed permeate pressure, in turn making data interpretation more straight forward.

Using the retentate gas composition and total pressure, the retentate H₂ partial pressure could be calculated. Since the permeate was only H₂, the total vacuum pressure was the same as the H₂ partial pressure. The vacuum pressure was measured after a right-angle elbow connected to the permeator shell, so its temperature was approxi-

mately ambient. Also this cold cathode gauge was calibrated for N₂ rather than the H₂ gas of the permeate. Thus, the following correction was applied to determine the permeate pressure at the membrane:

$$P_{\text{At Membrane}} = P_{\text{Measured}} K_{\text{Gas Factor}} \sqrt{\frac{T_{\text{At Membrane}}}{T_{\text{Cold Cathode Gauge}}}}$$

The cold cathode gauge manufacturer listed the gas factor as 0.43. No attempt was made to correct for conductance pressure losses.

4. Results

Table 1 is included as an example of the results collected. For this run the temperature was maintained at 450°C and the H₂ flowrate was fixed at 25 sccm. For all runs the retentate pressure was the vent pressure which was 590 Torr.

Table 1
Example results at 450°C and 25 sccm H₂ with various inert flow

Total feed flow rate (sccm)	Hydrogen partial pressure (Torr)	Measured permeate pressure (Torr)	Retentate concentration (ppm)	Feed P (Torr)	Corrected permeate pressure (Torr)	Retentate hydrogen partial pressure (Torr)
50	315.8	0.00068	10.1	628	0.00241	0.00598
100	157.3	0.00068	8.8	628	0.00241	0.00525
200	78.3	0.00068	9.0	630	0.00241	0.00534
300	53.1	0.00068	9.0	637	0.00241	0.00535
325	45.6	0.00068	8.9	637	0.00241	0.00529
350	46.3	0.00068	9.2	637	0.00241	0.00546
375	42.6	0.00068	11.8	640	0.00241	0.00697
400	40.0	0.00068	17.9	642	0.00241	0.01061
425	37.6	0.00068	33.3	645	0.00241	0.01970
450	35.5	0.00068	70.1	645	0.00241	0.04140
475	33.6	0.00068	130.4	645	0.00241	0.07695
500	31.4	0.00068	194.8	645	0.00241	0.11497

Fig. 3 shows results at 450°C and 5 sccm H₂. The retentate and permeate H₂ partial pressures are plotted against the total feed flowrate (5 sccm H₂ + inert). As shown, at all flowrates tested, the retentate and permeate partial pressures are approximately the same. Within experimental uncertainties, these values may be considered to be insubstantially different.

Fig. 4 shows the same parameters measured at 450°C and 25 sccm H₂. Compared with the 5 sccm case, the permeate pressure has increased due to the increased load on the turbo pump. At the lower total flowrates, again, the permeate and retentate partial pressures appear to be substantially the same. However, now at about 375 sccm total the retentate partial pressure begins to rise significantly above the permeate pressure. This point is referred to as ‘breakthrough’. At higher flowrates, ever-increasing amounts of hydrogen pass through the permeator and into the retentate stream.

Fig. 5 plots the retentate and permeate pressures for 450°C and 50 sccm H₂. The results are

similar in character. The permeate pressure increases further due to the high load on the turbomolecular pump and the breakthrough point is at a lower total flowrate of about 145 sccm.

Fig. 6 are the only data collected at 350°C where the H₂ flowrate was 25 sccm. In this case breakthrough was observed, and it was at about 265 sccm total.

Fig. 7 is a somewhat busy plot showing all 400°C data, which included H₂ flowrates of 1, 5, 25, 50 and 75 sccm. Breakthrough was not observed for 1 and 5 sccm H₂, and no H₂ was detected in the retentate (i.e. less than 0.3 ppm). For 25 sccm H₂ the lower flowrate data showed no detectible H₂ followed by breakthrough at about 330 sccm total. At 50 and 75 sccm H₂, hydrogen was detected at all conditions tested and breakthrough was observed at about 205 and 177 sccm total, respectively.

Table 2 is a summary of the temperature and H₂ flowrate conditions tested and the amount of inert required for breakthrough.

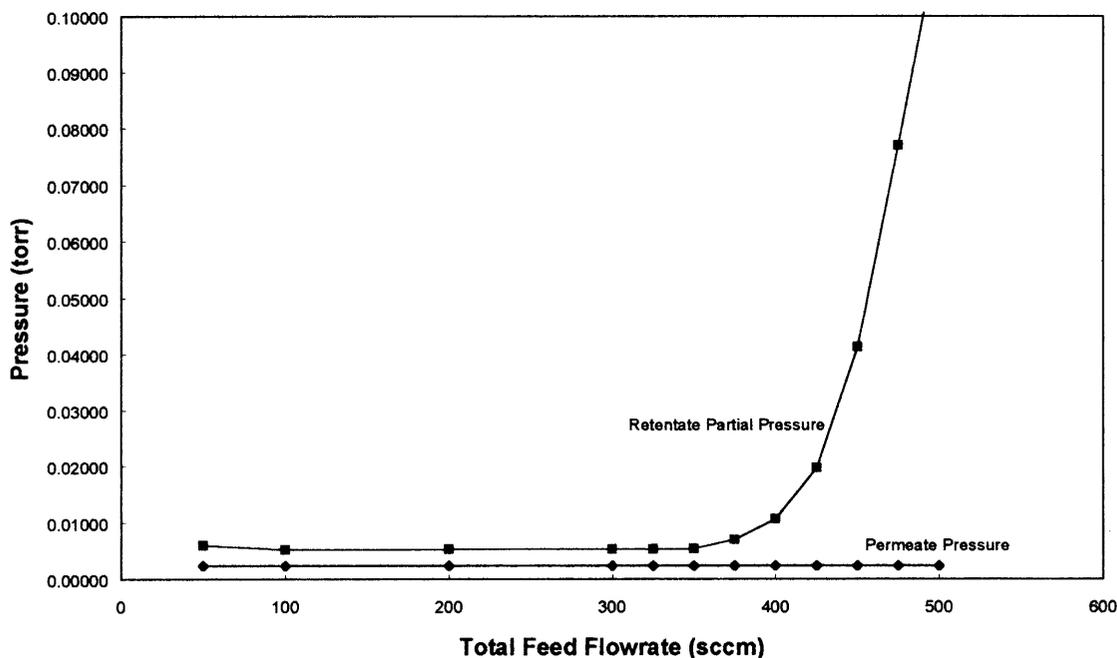
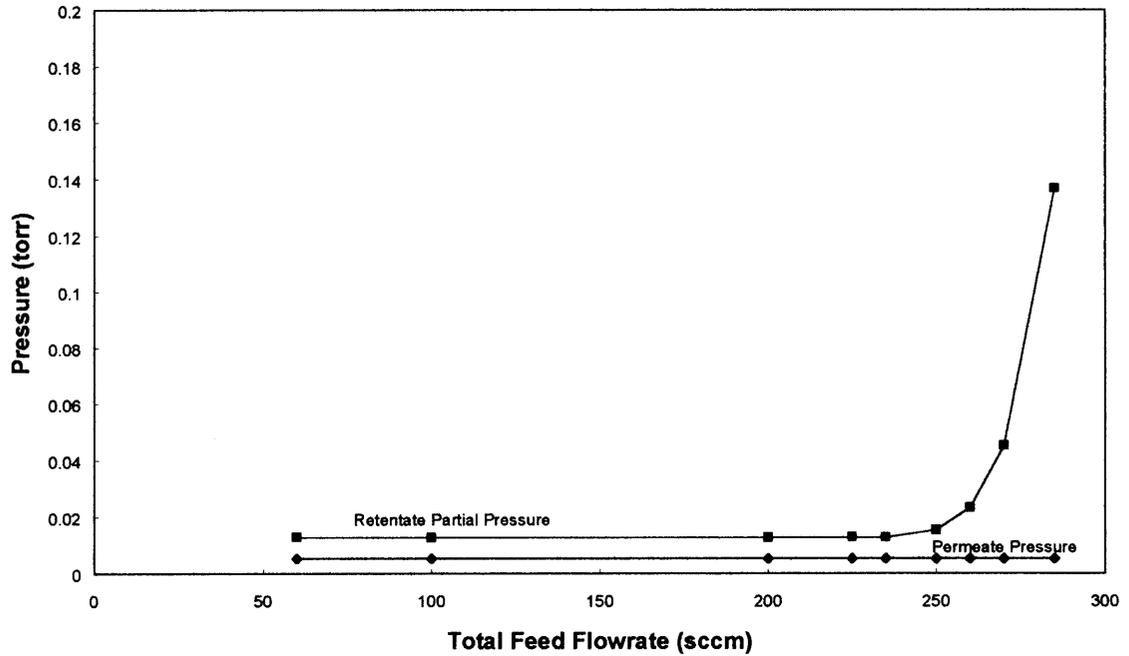
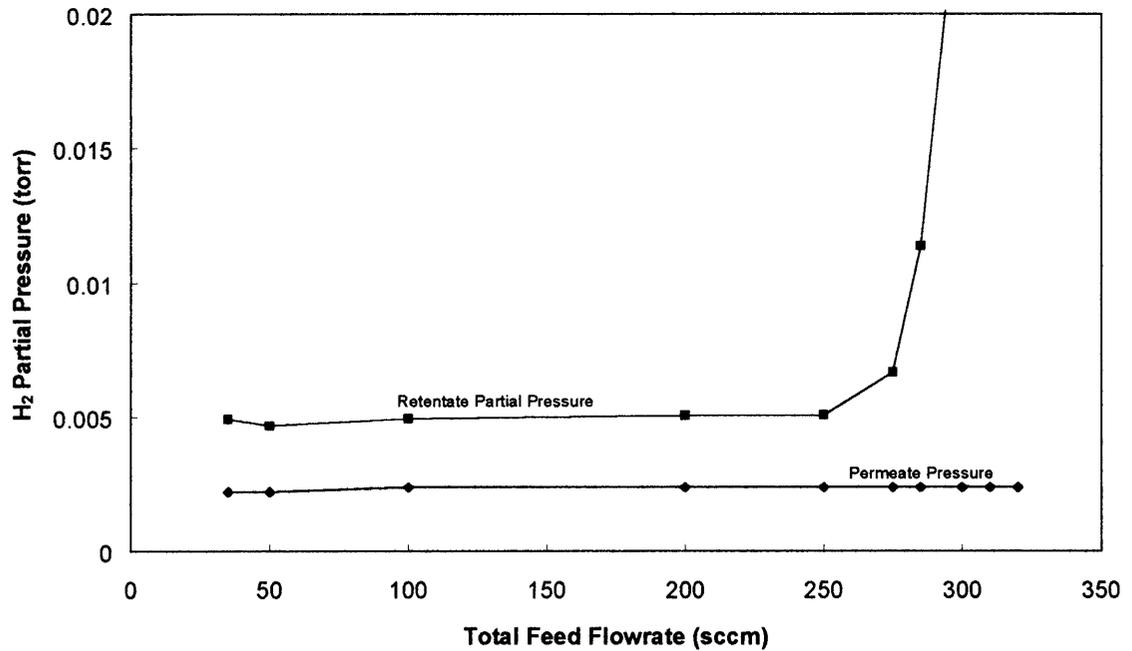


Fig. 4. Results at 450°C and 25 sccm H₂.

Fig. 5. Results at 450°C and 50 sccm H₂.Fig. 6. Results at 350°C and 25 sccm H₂.

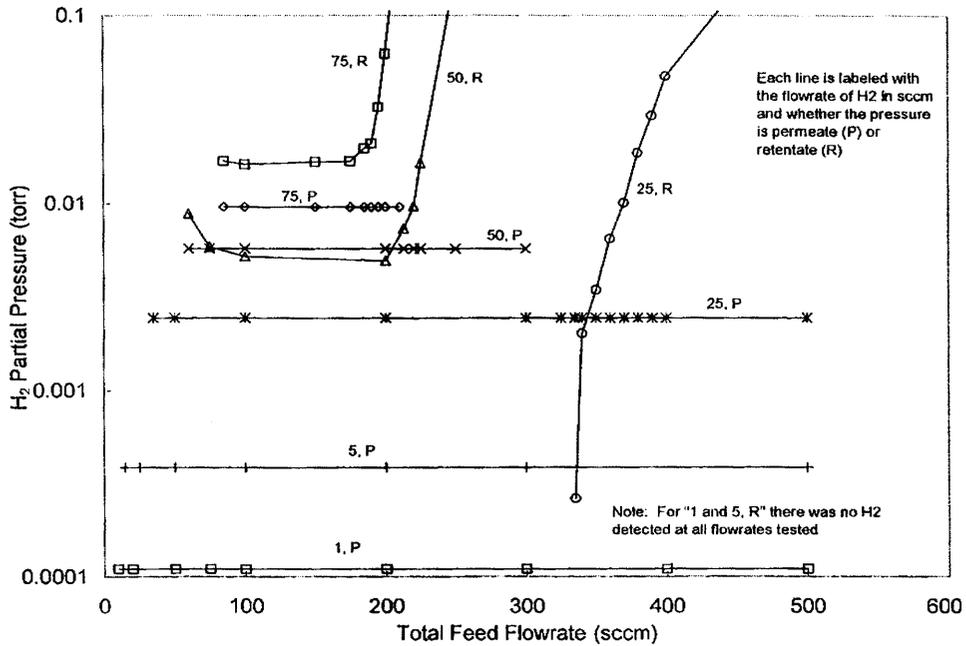


Fig. 7. Results at 400°C and H₂ flowrates from 1 to 75 sccm.

5. Discussion

It is known that rate-controlling mechanisms for this type of permeation differ at higher and lower pressures (permeation and dissociation/recombination). Prior to these experiments there was concern that the approach to partial pressure equilibrium might become excruciatingly slow when this low-pressure region was used. Rather, the results showed that partial pressure equilibrium was achieved at quite substantial flowrates in a small permeator. This lays a solid basis for using permeators for practical applications in this low-pressure region. For instance, a system such as the one shown in Fig. 8 can be envisioned for recovery of trace tritium from inert gas. As shown, the tritium is recovered in the permeator directly as T₂ and sent to storage for subsequent use. The retentate gas is sent to the stack through an oxidation/adsorption system to prevent any tritiated species such as water and methane from being directly vented.

6. Conclusions

These exploratory experiments found that it is practical to consider using Pd/Ag permeators for separating trace hydrogen isotopes from inert gases so that the remaining inert gases can be

Table 2
Summary of breakthrough conditions

T (°C)	F_{H_2} (sccm)	F_{inert} (sccm)
350	25	240
400	1	>499
400	5	>495
400	25	305
400	50	155
400	75	102
450	5	>495
450	25	350
450	50	195

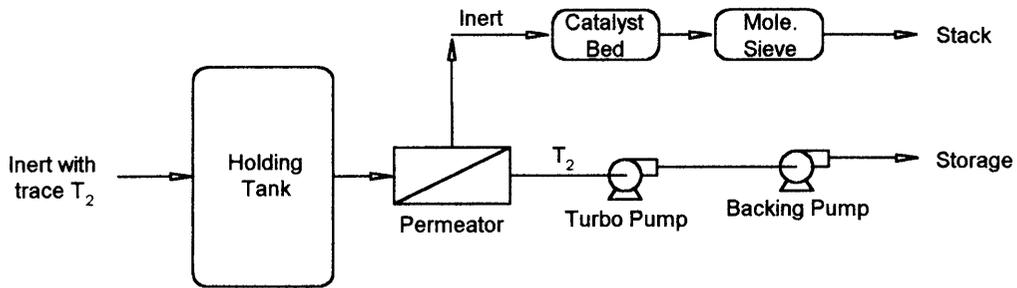


Fig. 8. Proposed system for recovery of trace tritium from inert gas.

released directly to the environment. It was found that a small, single-tube permeator achieved partial pressure equilibrium at flowrates at up to 500 sccm total feed flowrate. Results such as these were observed at all three temperatures tested, 350, 400 and 450°C and in the permeate pressure range 10^{-2} – 10^{-4} Torr. Under higher feed flowrate conditions the permeator could be driven to breakthrough so that preliminary scaling information for future work is available. Based on

these results an overall system based on this concept was proposed.

References

- [1] S.A. Birdsell, R.S. Willms, R.C. Wilhelm, Ultra-high tritium decontamination of simulated fusion fuel exhaust using a 2-stage palladium membrane reactor, in: Proceedings of the ANS 12th Topical Meeting on Technology of Fusion Energy, Reno, June 16–20, 1996.